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Quantum-optical analysis of high-order harmonic generation

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Quantum-optical analysis of high-order harmonic generation

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Abstract. High-order harmonic generation is a highly nonlinear phenomenon in which ultraviolet photons are generated when an atom is subjected to a high-intensity infrared laser field. This process has been extensively studied, but most of the formulations employ a semiclassical approach in which the electron is treated quantum mechanically while the driving field is treated classically. Although some recent studies follow a quantum-optical approach, the existing descriptions do not offer much insight into the physical processes taking place, and are always limited to monochromatic driving fields. In this thesis, we construct a fully quantized framework for strong-field ionization and high-harmonic generation that allows for quantization of both the driving field and the ultraviolet emission. The presented formalism extends the Strong-Field Approximation theory to a quantum-optical framework, with which we can obtain an analytical expression for the electron and the driving field after the interaction.

Keywords: High-Order Harmonic Generation, Quantum Optics, Strong-Field Physics.

1. Introduction

The motion of an electron within an atom takes place in attosecond time scales, so if we want to study and perform measurements in that time domain, then we need to use attosecond pulses [1,2]. In this framework, high-order harmonic generation is one of the most important phenomena and it is widely used, together with other amplitude and phase controlling techniques [3], since it gives rise to XUV radiation using high intensity infrared light sources.

High-order harmonic generation (HHG) is a highly nonlinear process in which an ionized electron interferes with the atomic bound state that it left behind, generating XUV radiation as a consequence. The main characteristics of this phenomenon are that the generated light comes as odd multiples of the introduced frequency (which range from NIR to MIR frequencies), forming an intensity plateau that breaks up at the so-called *cutoff frequency* given, in atomic units, by $\Omega_{\max} = I_p + 3.17U_p$ with I_p the ionization potential and U_p the ponderomotive energy, that is, the average kinetic energy of the oscillations of a free electron in a laser field [4].

This process was first seen experimentally, within gases, in 1987 [5], and an explanation covering the physics was provided by the so-called simple man's model or three-step model [2]:

- **Ionization.** The strong field applied to the target atom modulates the atomic potential forming a potential-energy barrier that an electron can tunnel out to reach the continuum.
- **Acceleration.** Once in the continuum, the field accelerates the liberated electron, increasing its kinetic energy and driving it back to the atom.

- **Recombination.** When the field drives back the electron to the atom, a recollision process can take place in which the kinetic energy gained by the electron is liberated in the form of high-energy photons.

Phenomena arising from strong-field ionization has been exhaustively studied from a semiclassical framework, in which electrons are treated quantum mechanically while the field is treated as a classical quantity. In this context, especially relevant is the Strong-Field Approximation (SFA) theory, which can be described as the study of light-matter interaction in regimes where the light's intensity is so strong that the perturbative analysis with respect to the electromagnetic field becomes invalid. It was first used by L. V. Keldysh in the study of multi-photon ionization processes [7], and later on extended to the analysis of high-order harmonic generation phenomena [8]. On the other hand, numerical methods based on 3D analysis of the time-dependent Schrödinger equation [6] have also become very useful in the study of these processes.

Nevertheless, despite some early work [9], it is only recently that attention has been turned to the fully quantized regime, in which both electron and driving field are treated quantum mechanically. In particular, novel work has developed SFA quantum-mechanical interpretations in terms of quantum information theory of the recombination process [10], and provided a quantized-IR version of the Volkov wavefunctions [11] that permits the calculation of quantum optical signatures in the final state of the input field. Moreover, these effects have been seen experimentally in gas-phase [12] and solid-state systems [13]. In another direction, there are also analyses which quantize the XUV modes, working numerically with a classical IR driver [14,15]. However, the existing work on quantized-field HHG does not offer too much insight into the physical processes taking place, and the discussions are constrained to single-mode fields, which does not allow the inclusion of effects arising from the shape of the pulse's envelope.

In this thesis we propose a complete quantized approach, i.e., one in which both IR and XUV are quantized, to strong-field ionization and high-order harmonic generation based on the Strong-Field Approximation. This approach gives a clear picture of those processes, which lead to an entangled state for the electron and the driving field, and it can be extended straightforwardly to a multimode situation. In this direction, we obtain a quantized version of the Volkov propagator that allows us to propose a quantum-optical SFA. We then employ these results to evaluate two different physical observables: the HHG spectrum, which coincides with the semiclassical spectrum, and the characteristic function associated to the Wigner distribution for above-threshold ionization, an experimentally measurable quantity in which quantum-optical signatures of strong-field processes can be measured.

This report is structured as follows: in Sec. 2 we introduce some transformations and approximations over the Hamiltonian of a free-electron travelling in a single-mode quantized field, which allow us to simplify later expressions. In Sec. 3 we obtain a full quantum version of the Volkov propagator for an extension of the previous Hamiltonian to a multimode situation, using a method known as Wei-Norman factorization [16]. In Sec. 4 we use strong-field assumptions to get an expression, written in terms of Magnus expansions [17], that contains information about ionization and HHG phenomena. In Sec. 5 we use the previous solution to get an expression for the HHG spectrum that can be compared with the semiclassical result, and in Sec. 6 we calculate the characteristic function associated to the Wigner distribution for above-threshold ionization. Finally, in Sec. 7 we summarise the results obtained and comment on possible directions for future work. Moreover, this report is accompanied by a Supplementary Material (Secs. S1-S13) that contains explicit derivations of the results in this work, and further discussions of the approximations used.

2. Free electron Hamiltonian in a quantized field

In the velocity-gauge minimal-coupling, employing atomic units and working under the dipole approximation, the Hamiltonian of a free electron in a single-mode quantized field of frequency ω_0

reads [18]

$$H = \frac{1}{2}\big[\hat{P} + \hat{A}\big]^2 + \omega_0 \hat{b}^\dagger \hat{b} \quad \text{with} \quad \hat{A} = A_0\big[\hat{b} + \hat{b}^\dagger\big], \tag{2.1}$$

where $\hat{P}$ is the canonical momentum operator; $\hat{b}^\dagger$ and $\hat{b}$ are, respectively, the creation and annihilation operators acting over the considered mode of the applied field; and $\hat{A}$ the vector potential with $A_0 = c\sqrt{\pi/\omega_0 V}$ the vector's potential amplitude [19] that depends on the light's velocity c and the quantization volume V.

Typically, the lasers that are used in these experiments have a pulse duration of 30 fs, a beam width of approximately 1 to 50 μm and a frequency between 800 nm to 2.4 μm, which leads to values of A_0 ranging from 10^{-6} to 10^{-8}, given in atomic units. Therefore, in our analysis we will neglect terms proportional to A_0^2 but preserve those linear in A_0. However, we are also working in regimes where the number of photons N is a very large quantity but requiring that $N/V \sim$ constant.

In the absence of electron interactions with the photonic state, the latter describes a counterclockwise rotation in the corresponding phase space, so that its state is characterized by

$$|\psi_0(t)\rangle_{\text{field}} = \left|\alpha e^{-i\omega_0 t}\right\rangle, \tag{2.2}$$

Let us introduce a set of transformations and approximations that simplifies the analysis in the photonic phase space and that provides a clear picture which we can identify with the semiclassical one. All the calculations shown in this section are explicitly developed in Sec. S2.

(i) *Interaction picture with respect to the free field term.* Eqs. (2.1) and (2.2), are formulated in the Schrödinger picture, in which quantum states evolve in time while operators remain unchanged. This transformation reverses that situation, i.e., stops the motion of the coherent state in the associated phase-space, and transfer all that time dependence to the vector potential, described from this point forward by $\hat{A}(t)$, so that it contains the field's time evolution. In other words, this transformation makes the photonic phase space frame of reference to rotate with the same frequency ω_0 as the coherent state, so this latter one is seen as if it was at rest:

$$|\psi(t)\rangle = e^{-i\omega_0 \hat{b}^\dagger \hat{b} t}\,|\psi_{\text{I}}(t)\rangle \Rightarrow H_{\text{I}}(t) = \frac{1}{2}\big[\hat{P} + \hat{A}(t)\big]^2 \quad \text{with} \quad |\psi_{\text{I}}(t_0)\rangle_{\text{field}} = |\alpha\rangle\,, \tag{2.3}$$

where

$$\hat{A}(t) = A_0\big[\hat{b}e^{-i\omega_0 t} + \hat{b}^\dagger e^{i\omega_0 t}\big]. \tag{2.4}$$

(ii) *Photonic displacement.* This transformation consists on a displacement in the photonic phase space of a quantity α, i.e., we apply the displacement operator $D(\alpha)$ [19], so that in this new frame of reference we can split the vector potential in two contributions:

$$|\psi_{\text{I}}(t)\rangle = D(\alpha)\,|\psi_{\text{II}}(t)\rangle \Rightarrow H_{\text{II}}(t) = \frac{1}{2}\Big[\hat{P} + A(t) + \delta\hat{A}(t)\Big]^2 \quad \text{with} \quad |\psi_{\text{II}}(t_0)\rangle_{\text{field}} = |0\rangle\,. \tag{2.5}$$

Those contributions of the vector potential are $A(t)$, the mean value of the vector potential operator over the initial vacuum state, which acts as the classical vector potential

$$A(t) = A_0\big[\alpha e^{-i\omega_0 t} + \alpha^* e^{i\omega_0 t}\big], \tag{2.6}$$

and $\delta\hat{A}(t)$ which describes quantum fluctuations

$$\delta\hat{A}(t) = A_0\big[\hat{b}e^{-i\omega_0 t} + \hat{b}^\dagger e^{i\omega_0 t}\big] = \sqrt{2}A_0\big[\hat{X}_\varphi \cos(\omega_0 t + \varphi) + \hat{X}_{\bar{\varphi}} \sin(\omega_0 t + \varphi)\big]. \tag{2.7}$$

Here $\hat{X}_\varphi$ and $\hat{X}_{\bar{\varphi}}$ are quadrature operators at phases φ and $\bar{\varphi} - \pi/2$:

$$\hat{X}_\varphi = \frac{\hat{b}e^{i\varphi} + \hat{b}^\dagger e^{-i\varphi}}{\sqrt{2}}, \quad \hat{X}_{\bar{\varphi}} = \frac{\hat{b}e^{i\bar{\varphi}} + \hat{b}^\dagger e^{-i\bar{\varphi}}}{\sqrt{2}}, \text{ and they obey } \left[\hat{X}_\varphi, \hat{X}_{\bar{\varphi}}\right] = i. \tag{2.8}$$

Then, if we neglect at this point quadratic terms in A_0^2, our final velocity gauge Hamiltonian $H_V(t)$ reads

$$H_V(t) \approx \frac{1}{2}\Big\{\big[\hat{P} + A(t)\big]^2 + 2\big[\hat{P} + A(t)\big]\,\delta\hat{A}(t)\Big\}. \tag{2.9}$$

(iii) *Semiclassical length gauge transformation.* This transformation is the one used in the semiclassical studies [19] to go from the velocity gauge to the length gauge, reason why we call it *semiclassical*. It acts only over electronic states and allow us to change the canonical momentum p by the kinetic momentum $k(t) = p + A(t)$

$$|\psi_{\text{II}}(t)\rangle = e^{i\hat{X}A(t)}\,|\psi_L(t)\rangle \Rightarrow H_L(t) = \frac{1}{2}\hat{P}^2 + \hat{X}F(t) + \hat{P}\,\delta\hat{A}(t), \text{ with } F(t) = -\frac{\partial A(t)}{\partial t}, \tag{2.10}$$

with $F(t)$ the field's amplitude. Here it is important to remark that $\hat{P}$ now represents the electron's kinetic momentum which now coincides with the canonical momentum.

The extension to the multimode case follows straightforwardly from this point, where eq. (2.9) remains unchanged but with

$$\delta\hat{A}(t) = \sum_\omega A_\omega\big[\hat{b}_\omega e^{-i\omega t} + \hat{b}_\omega^\dagger e^{i\omega t}\big] \text{ with } A_\omega = c\sqrt{\pi/\omega V}, \tag{2.11}$$

where $\hat{b}_\omega$ and $\hat{b}_\omega^\dagger$ are the creation and annihilation operators acting over the mode of frequency ω.

3. Quantized Volkov propagator

In this section we study the dynamics of a free electron interacting with a quantized electromagnetic field. With that purpose, we are going to employ a method which is known as *Wei-Norman factorization* [16], and that provides a solution to the differential equation

$$i\frac{\partial U(t)}{\partial t} = H(t)U(t) \text{ with } H(t) = \sum_{i=1}^{n} a_i(t)\hat{Z}_i \text{ and } U(t_0) = \mathbb{1}, \tag{3.1}$$

where the $a_i(t)$ are some time-dependent functions and the $\hat{Z}_i$ belong to a set of generators of an $\mathcal{N}$-dimensional Lie algebra. A solution to this differential equation is given by

$$U(t) = \prod_{i=1}^{\mathcal{N}} e^{g_i(t)\hat{Z}_i} = e^{g_1(t)\hat{Z}_1}e^{g_2(t)\hat{Z}_2}...e^{g_{\mathcal{N}}(t)\hat{Z}_{\mathcal{N}}}, \tag{3.2}$$

where $\left\{\hat{Z}_{n+1}, ..., \hat{Z}_{\mathcal{N}}\right\}$ are the operators needed to complete the previous algebra and the $g_i(t)$ functions are determined by

$$i\sum_{l=1}^{\mathcal{N}}\sum_{j=1}^{\mathcal{N}} \dot{g}_l(t)\zeta_j(t)\hat{Z}_j = \sum_{i=1}^{n} a_i(t)\hat{Z}_i, \tag{3.3}$$

with $\zeta_i(t) = \zeta_i(g_1(t), ..., g_{\mathcal{N}}(t))$ functions that arise from the evaluation of the so-called *Wei-Norman operator* (see S3).

For a multimode situation, we can check by examination of all the possible commutators that the following set of operators form a closed set under commutation and, thus, a Lie algebra

$$\{\mathbb{1}, \hat{P}, \hat{P}^2\} \cup \{\hat{X}_{\varphi,\omega},\ \hat{X}_{\bar{\varphi},\omega},\ \hat{P}\hat{X}_{\varphi,\omega},\ \hat{P}\hat{X}_{\bar{\varphi},\omega} : \omega \in \mathbb{R}^+\}. \tag{3.4}$$

Under the transformations that we have considered until now, the Wei-Norman factorization seems especially useful since the only operators that do not commute are the ones that involve different quadratures of the same photonic phase space. This is something that does not happen if either the diamagnetic term is considered or an analysis within the length gauge is done (see S4), and a set of differential equations that can be solved analytically, like the ones shown below (see S3), does not necessarily appear

$$\begin{cases} i\dot{g}_{\hat{P}^2} + \sum_\omega \dot{g}_{\hat{P}\hat{X}_{\varphi,\omega}} g_{\hat{P}\hat{X}_{\bar{\varphi},\omega}} = \frac{1}{2} \\ i\dot{g}_{\hat{P}} + \sum_\omega \left[\dot{g}_{\hat{P}\hat{X}_{\varphi,\omega}} g_{\hat{X}_{\bar{\varphi},\omega}} + \dot{g}_{\hat{X}_{\varphi,\omega}} g_{\hat{P}\hat{X}_{\bar{\varphi},\omega}}\right] = A(t) \\ i\dot{g}_{\mathbb{1}} + \sum_\omega \dot{g}_{\hat{X}_{\varphi,\omega}} g_{\hat{X}_{\bar{\varphi},\omega}} = \frac{1}{2}A^2(t) \end{cases}, \quad \begin{cases} i\dot{g}_{\hat{P}\hat{X}_{\bar{\varphi},\omega}} = \sqrt{2}A_\omega \sin(\omega t + \varphi_\omega) \\ i\dot{g}_{\hat{X}_{\bar{\varphi},\omega}} = \sqrt{2}A_\omega A(t) \sin(\omega t + \varphi_\omega) \\ i\dot{g}_{\hat{P}\hat{X}_{\varphi,\omega}} = \sqrt{2}A_\omega \cos(\omega t + \varphi_\omega) \\ i\dot{g}_{\hat{X}_{\varphi,\omega}} = \sqrt{2}A_\omega A(t) \cos(\omega t + \varphi_\omega) \end{cases}, \tag{3.5}$$

which lead, after some operations (see S5 for a single-mode case discussion) to the quantized Volkov operator

$$U_V(t,t') = e^{-\frac{i}{2}\int_{t'}^{t}[\hat{P}+A(t)]^2} \mathcal{D}(\hat{P},t,t') \quad \text{with} \quad \mathcal{D}(\hat{P},t,t') = \prod_\omega D_\omega\big(\alpha_\omega(\hat{P},t,t')\big), \tag{3.6}$$

and $\alpha_\omega(\hat{P},t,t')$ defined as

$$\alpha_\omega(\hat{P},t,t') = -iA_\omega \int_{t'}^{t} \mathrm{d}\tau \big[\hat{P} + A(\tau)\big] e^{i\omega\tau}. \tag{3.7}$$

This result has a lot of valuable physical information: it tells us that when a free electron interacts with an electromagnetic field, the total state of the system gets modified by a phase that coincides with the semiclassical action [20], but also is affected by an operator which introduces a two-mode squeezing that mixes electronic modes with the photonic mode of frequency ω. This operation has very important physical implications, as it entangles the field with the photonic modes something that affects ionization and HHG phenomena and that we discuss in Sec. 4.

Besides, the extension of this operator to the length gauge is obtained from eq. (3.6) as follows

$$U_L(t,t') = e^{i\hat{X}A(t)} U_V(t,t') e^{-i\hat{X}A(t')}, \tag{3.8}$$

where first, we move to the velocity gauge at time t', then the system evolves in time from t' to t within that gauge through $U_V(t,t')$, and finally we return to the length gauge at time t.

4. Quantum-Optical Strong-Field Approximation

The differential equation that dictates the dynamics of ionization and HHG processes, under the transformations and approximations we have introduced thus far, is given by

$$i\frac{\partial U(t)}{\partial t} = \left[\frac{\hat{P}^2}{2} + \hat{V}_{\text{at}} + \hat{X}F(t) + \hat{P}\,\delta\hat{A}_{\text{ir}}(t) + \hat{P}\,\delta\hat{A}_{\text{uv}}(t)\right] U(t), \tag{4.1}$$

where $\hat{V}_{\text{at}}$ is the atomic potential. Note that we have divided quantum fluctuations $\delta\hat{A}(t)$ in two terms: $\delta\hat{A}_{\text{ir}}(t)$ that incorporates the field modes, and $\delta\hat{A}_{\text{uv}}(t)$ which includes all the other modes,

in particular the XUV modes generated in HHG. This division is another fundamental point of the present approach, thanks to the fact that a solution to the previous differential equation can be written in terms of a Magnus expansion, also known in the literature as Dyson expansion [21]

$$U(T,t_0) = U_0(T,t_0) - i\int_{t_0}^{T} \mathrm{d}t'\; U(T,t')\hat{V}(t')U_0(t',t_0), \tag{4.2}$$

where the partition is $H(t) = H_0(t) + \hat{V}(t)$, with $U_0(t,t')$ the propagator for the first term and with $U(T,t')$ satisfying eq. (4.1) (see S6).

At this point, we apply the Magnus expansion twice using a different partition each time. For the first one, we split the Hamiltonian as follows

$$H(t) = H_{\mathrm{at}} + \hat{V}_{\mathrm{int}}(t), \text{ with } \hat{V}_{\mathrm{int}}(t) = \hat{X}F(t) + \hat{P}\,\delta\hat{A}_{\mathrm{ir}}(t) + \hat{P}\,\delta\hat{A}_{\mathrm{uv}}(t) \tag{4.3}$$

obtaining by means of eq. (4.2)

$$U(T,t_0) = U_{\mathrm{at}}(T,t_0) - i\int_{t_0}^{T} \mathrm{d}t'\; U(T,t')\hat{V}_{\mathrm{int}}(t')U_{\mathrm{at}}(t',t_0), \tag{4.4}$$

where $U_{\mathrm{at}}(T,t_0)$ is the propagator of H_{at}, the atomic Hamiltonian.

If at this point we take the limit in which quantum fluctuations tend to zero, i.e., $A_\omega \to 0\ \ \forall\omega$, then $V_{\mathrm{int}}(t) = \hat{X}F(t)$ and the semiclassical solution [8] is recovered. In this way, the first term of eq. (4.4) gives the probability of not having any interaction with the field, while the integral term incorporates ionization processes through $\hat{X}F(t)$, and other phenomena (like spontaneous emission – see S9 – through the quantum fluctuations). Nevertheless, we shall follow a different route by introducing another Magnus expansion for the propagator $U(T,t')$ that appears inside the integral, now using a different partition given by

$$H(t) = H_{\mathrm{ir}}(t) + \hat{V}_{\mathrm{uv}}(t), \text{ with } H_{\mathrm{ir}}(t) = H_{\mathrm{at}} + \hat{P}\,\delta\hat{A}_{\mathrm{ir}}(t) \text{ and } \hat{V}_{\mathrm{uv}}(t) = \hat{P}\,\delta\hat{A}_{\mathrm{uv}}(t), \tag{4.5}$$

which leads to the following solution

$$\begin{aligned} U(T,t_0) = U_0(T,t_0) &- i\int_{t_0}^{T} \mathrm{d}t'\; U_{\mathrm{ir}}(T,t')\hat{V}_{\mathrm{int}}(t')U_0(t',t_0) \\ &- \int_{t_0}^{T}\mathrm{d}t' \int_{t'}^{T}\mathrm{d}t\; U(T,t)\hat{V}_{\mathrm{uv}}(t)U_{\mathrm{ir}}(t,t')\hat{V}_{\mathrm{int}}(t')U_0(t',t_0), \end{aligned} \tag{4.6}$$

where $U_{\mathrm{ir}}(T,t')$ is the field-entangled propagator for $H_{\mathrm{ir}}(t)$. Note that by following this partition, we obtain a third term in the expression of $U(T,t_0)$ which incorporates an electron-field interaction at time t' through $\hat{V}_{\mathrm{int}}(t')$, then introduces an entangled evolution in time of the electron and the modes belonging to the field through $U_{\mathrm{ir}}(t,t')$ until t, moment at which another extra interaction is added through $\hat{V}_{\mathrm{uv}}(t)$, operator that allows us to introduce XUV photons in the discussion, in agreement with the definition of $\delta\hat{A}(t)$ shown in eq. (2.11), that were not initially in the input field. Therefore, we will identify this term, later on in this section, with the one describing recombination phenomena after the acceleration and ionization steps.

To clear up the physics in eq. (4.6), we will consider the initial state of the system to be

$$|\Psi(t_0)\rangle = |\epsilon_0, \{0\}\rangle \equiv |\mathrm{g}, \{0\}\rangle \tag{4.7}$$

where $\{0\}$ indicates that, initially, all the field modes are in vaccum states. Then we introduce the completeness relation in the atomic basis, i.e.,

$$\mathbb{1} = \sum_n |\epsilon_n\rangle\langle\epsilon_n| + \int \mathrm{d}k\, |\phi_k\rangle\langle\phi_k|\,, \text{ with } \hat{H}_{\mathrm{at}}\,|\epsilon_n\rangle = -|\epsilon_n|\,|\epsilon_n\rangle \text{ and } \hat{H}_{\mathrm{at}}\,|\phi_k\rangle = E_{\phi_k}\,|\phi_k\rangle\,, \tag{4.8}$$

after each interaction potential of the form $\hat{V}(t)$ (see S7), and impose strong-field assumptions [8, 20]:

- the applied laser field is very strong so we will neglect the atomic potential during the time evolution, that is, $U_{\rm ir}(t,t') \approx U_{\rm L}(t,t')$, where $U_{\rm L}(t,t')$ is the length gauge Volkov propagator introduced in eq. (3.8);
- we will neglect the contribution to the evolution of the system of all bound states except the ground state;
- we will approximate continuum states by Volkov states (further details in S8), i.e.,

$$\hat{P}\,|\phi_k\rangle = k\,|\phi_k\rangle \longrightarrow E_{\phi_k} = \frac{1}{2}k^2, \tag{4.9}$$

  where k is the kinetic momentum;
- we will neglect continuum-continuum transitions.

After all these operations which involve a lot of simplifications (see S7), we can work out the following Strong-Field quantized solution

$$\begin{aligned}|\Psi(T,t_0)\rangle &= U_{\rm at}(T,t_0)\,|{\rm g},\{0\}\rangle \\ &- i\int {\rm d}p \int_{t_0}^{T} {\rm d}t' e^{-iS_{\rm sc}(p,T,t')} F(t'){\rm d}\big(p+A(t')\big)\mathcal{D}(p,t,t')\,|p+A(T),\{0\}\rangle \\ &- \sum_{\Omega\in{\rm uv}} \int {\rm d}p \int_{t_0}^{T}{\rm d}t' \int_{t'}^{T} {\rm d}t\; e^{-iS(p,t,t')} G(p,t,t')\mathcal{D}(p,t,t')\,|{\rm g},\{0\},1_\Omega\rangle\,,\end{aligned} \tag{4.10}$$

where the explicit exponent term appearing in the second term is defined as

$$S_{\rm sc}(p,t,t') = \frac{1}{2}\int_{t'}^{t} {\rm d}\tau \big[p+A(\tau)\big]^2 - I_p(t'-t_0), \tag{4.11}$$

whereas the one appearing in the third term is

$$S(p,t,t') = -I_p(T-t) - \Omega t + S_{\rm sc}(p,t,t'). \tag{4.12}$$

On the other hand, the prefactor is determined by

$$G(p,t,t') = \left[\frac{(p+A(t))^2}{2} + I_p\right] F(t'){\rm d}^*\big(p+A(t)\big){\rm d}\big(p+A(t')\big). \tag{4.13}$$

The explicit exponents appearing in each of the terms of eq. (4.10) have a lot of valuable information about the physics, as they represent the electron's action. In the second term the appearing phase is $S_{\rm sc}(p,t,t')$, whose second term tells us that the electron stays in the ground state, i.e., with energy $-I_p$, until time t', and from there it acquires a momentum $p+A(t')$ that changes in time with the field, that is, the electron's kinetic energy increases because the field is making it to accelerate. This description coincides with the first two steps of the simple man's model and, therefore, this term represents ionization phenomena with t' the ionization time. On the other hand, the dynamics of the third term are described by $S(p,t,t')$ which also has ionization and acceleration in the continuum processes, but at time t it loses an amount of energy equal to Ω, falling again to the ground state. Therefore, we can identify time t with the recombination time and the lost energy Ω with an emitted photon, so that this third term describes HHG.

Furthermore, the displacement operator acting over the field modes also introduces an implicit exponent term, which we can obtain in the evaluation of physical observables. This extra term adds, to the electron's action, quantum effects generated during the acceleration step, as the field modes displace a quantity $\alpha_\omega(p,t,t')$ that makes them approach to a vacuum state along the $\hat{X}_{\varphi,\omega}$ quadrature, if we choose φ equal to the initial field's phase (see S10). Thus, this displacement represents absorption phenomena that take place throughout the whole acceleration step and that depends on the kinetic momentum acquired by the electron during that process.

5. High harmonic spectrum

As we have seen previously, the exponent that appears in the final state of the system gives information about the dynamics, as it represents the action; because of this, it will be the main focus of attention in the analysis of physical observables. In this section, those exponents will also incorporate the quantum fluctuation terms we discussed at the end of the previous section. Hence, to distinguish them from the ones shown in eqs. (4.11) and (4.12), we will call them $\mathcal{S}(p,t,t')$. Moreover, this quantity is a highly oscillatory function, that allows us to apply saddle point methods (see S11) in the resultant integrals. This method changes appropriately the integration contour, passing through saddle points of $\mathcal{S}(p,t,t')$, so that we can express our integrals as gaussian-like integrals. Those saddle points are determined by the so-called *saddle-point equation*

$$\partial\mathcal{S}(p_s,t_r,t_i)/\partial\eta = 0, \text{ where } \eta = \{p,t,t'\}. \tag{5.1}$$

with p_s the electron canonical momentum, t_i the ionization time and t_r the return time.

A way in which we can evaluate the HHG spectrum, in agreement with the semiclassical analysis, is by studying the Fourier transform of the electron dipole since the atomic polarization determines the response of an atom subjected to a field. In this way, we get the following total phase (see S12), which is the sum of the semiclassical action shown in eq. (4.12) [20] and an extra term that depends on how much energy from the photonic phase space has been absorbed

$$\mathcal{S} = S(p,t,t') - i\frac{|\alpha_{\text{ir}}(p,t,t')|^2}{2}. \tag{5.2}$$

This last term arises from the evaluation of a term whose form is $\langle 0|\alpha_{\text{ir}}(p,t,t')\rangle$, which is indeed the overlap between the photonic initial and final state: depending on their closeness, the spectrum will be suppressed or not. Indeed this is a similar result to the autocorrelation function that appears for HHG in the presence of nuclear motion in molecules [22, 23], with the main difference that the present autocorrelation takes place within the photonic sector.

Another more fundamental way in which we can calculate the HHG spectrum is through the analysis of the recombination term of eq. (4.10), that we will represent as $|\psi_{\text{r}}(T,t_0)\rangle$ and that is a superposition of states in which the electron returns to the ground state of the system while a photon of frequency Ω is generated, where Ω belongs to the XUV. Each of these states is multiplied by a certain amplitude that determines how probable a particular process is, and in consequence it will define the shape of the final recombination spectrum. Thus, we can write the probability of generating mode Ω in the recombination, up to normalization factors, as

$$P(\Omega) = \langle\Psi(T,t_0)|\hat{\Pi}_{1,\Omega}|\Psi(T,t_0)\rangle \text{ with } \hat{\Pi}_{1,\Omega} = |1_\Omega\rangle\langle 1_\Omega|\,, \tag{5.3}$$

and from here, the following expression for the total phase is obtained (see S12)

$$\begin{aligned}\mathcal{S} = S_2(p_2,t_2,t_2') - S_1(p_1,t_1,t_1') + \frac{1}{2i}\Big|\alpha_{\text{ir},2}(p_2,t_2,t_2') - \alpha_{\text{ir},1}(p_1,t_1,t_1')\Big|^2,\\ - \frac{1}{2i}\Big[\alpha_{\text{ir},1}(p_1,t_1,t_1')\alpha^*_{\text{ir},2}(p_2,t_2,t_2') - \alpha^*_{\text{ir},1}(p_1,t_1,t_1')\alpha_{\text{ir},2}(p_2,t_2,t_2')\Big],\end{aligned} \tag{5.4}$$

where the subscripts 1 and 2 distinguish the different integration variables that appear when we perform the mean value of $\hat{\Pi}_{1,\Omega}$.

There are similarities among eqs. (5.2) and (5.4), as quantum fluctuations come in both cases from projections between coherent states. Conversely, there are missing terms in the former since in the second equation the projection is taken between two non-null photonic coherent states.

However, these differences are negligible as quantum fluctuations are proportional to A_0^2, so the saddle points arising in both cases from $\mathcal{S}(p,t,t')$ will be almost equal between them and to the semiclassical ones. In conclusion, we obtain the same spectrum as the one arising from the semiclassical analysis.

6. Characteristic function for above-threshold ionization

Quantum signatures in the HHG spectrum are negligible, according to the calculations we showed in the previous section. Thus, the main question now is to find a physical observable for which these quantum fluctuations could affect the final saddle points and, in particular, we will try to find expressions for the action which are linear in the amplitude of the vector potential, i.e., A_0. A possible observable satisfying these requirements is the Wigner function, which in this section we study for the IR states at the ionization step. Besides, we can measure it experimentally by means of homodyne detection [19, 24].

In classical mechanics, the Liouville density determines the probability of finding a particle in a given point of the associated phase space [25]. The Wigner function [26] plays the same role as that density function but in the context of quantum mechanics, where new phenomena arising from the nature of the wavefunctions, like interference, appears. Due to this quantum nature, the Wigner function is said to be a *quasiprobability distribution* since it can be negative. On the other hand, as any classical random-variable probability function, it admits a representation in terms of the so-called characteristic function $\chi(z)$, defined as its Fourier transform, which contains the same information as the probability density function but in a different basis.

The characteristic function of a system described by the density matrix $\hat{\rho}$, is given by [27]

$$\chi(z) = \mathrm{tr}\big[\hat{\rho}\hat{W}_z\big], \tag{6.1}$$

with $\hat{W}_z$ the Weyl operator, which can be written as (see S13)

$$\hat{W}_z = D(z) \text{ and } z \equiv [x_2\cos(\varphi) - x_1\sin(\varphi)] - i[x_2\sin(\varphi) + x_1\cos(\varphi)] \tag{6.2}$$

where x_1 and x_2 are coordinates of a phase space point. In this case, our density matrix reads

$$\hat{\rho}_{\mathrm{ir},p} = |\psi_{\mathrm{ir},p}(T,t_0)\rangle\langle\psi_{\mathrm{ir},p}(T,t_0)|\,, \tag{6.3}$$

with $|\psi_{\mathrm{ir},p}(t,t_0)\rangle$ the ionization states (second term of eq. (4.10)) conditioned to the measurement of an electron with momentum $p + A(t)$, i.e., if we write eq. (4.10) as

$$|\Psi(T)\rangle = |\psi_{\mathrm{g}}(T)\rangle + |\psi_{\mathrm{ion}}(T)\rangle + |\psi_{\mathrm{r}}(T)\rangle\,,\ \text{then}\ \ |\psi_{\mathrm{ir},p}(T)\rangle = \langle p + A(T)|\psi_{\mathrm{ion}}(T)\rangle\,. \tag{6.4}$$

After some calculations (detailed in S13), we get for the characteristic function

$$\chi(z) = \int_{t_0}^{t}\mathrm{d}t_1'\int_{t_0}^{t}\mathrm{d}t_2' F(t_1')F(t_2')\mathrm{d}^*\big(p + A(t_1')\big)\mathrm{d}\big(p + A(t_2')\big)e^{i\mathcal{S}}, \tag{6.5}$$

where the exponent $\mathcal{S}$, after writing $|z| = ze^{-i\theta}$ and neglecting terms proportional to A_0^2, is

$$\begin{aligned}\mathcal{S} &\approx S_{\mathrm{sc},1} - S_{\mathrm{sc},2} + i\frac{|z|^2}{2}\\ &+ A_0\sum_{j=1}^{2}\left[|z|\int_{t_j'}^{t}\mathrm{d}\tau_j[p + A(\tau_j)]\cos(\omega\tau_j + \theta) + i(-1)^j|z|\int_{t_j'}^{t}\mathrm{d}\tau_j[p + A(\tau_j)]\sin(\omega\tau_j + \theta)\right],\end{aligned} \tag{6.6}$$

As we can see, this total phase presents terms which are linear in A_0, and this then implies that the Wigner function is a possible physical observable that may allow the measurement of quantum effects during strong-field ionization. According to the semiclassical analysis, the final state for the photonic part remains unchanged so a gaussian behaviour should be expected for a probability density function arising from the fact we have an unchanged coherent state. On the other hand, the effects introduced when we consider the quantum nature of the field are still small, but not as much as in the HHG spectrum so in principle, as $\mathcal{S}$ is not symmetric upon the change $t_1 \leftrightarrow t_2$ since the cosine term does not change its sign, the saddle points will be slightly different and, furthermore, will affect the final Wigner function since the terms proportional to A_0 appear together with a z term, so that the final quasiprobability distribution would be slightly different from a gaussian, affecting this fact to the final Fourier transform.

7. Conclusions

In this work, we have developed a fully quantized approach to strong-field ionization and high-order harmonic generation based on Strong-Field Approximation, which naturally extends to a multimode construction and that gives a clear picture about the underlying physics.

Apart from agreeing with the three-step model, this proposal provides us with information about the photonic phase space. First, it tells us that during the acceleration step the dynamics of the system are described by a two-mode squeezing operator that mixes the electronic modes with the field photonic modes, in such a way that the initial state of the system, which can be written as separable state, becomes entangled: depending on the momentum acquired by the electron, the coherent state that characterizes the field displaces a certain amount in the corresponding phase space, where we can identify this displacement with absorption phenomena since it moves the coherent state towards the initial origin, that is, to a vacuum state. Hence, the final state of the system is given as a superposition of all the possible displacements.

In the second place, this proposal tells us that the photon states arising from HHG are single-photon Fock states, at least for a single-atom emission. However, we know that Fock states have a completely undefined phase, so this fact opens a very interesting question regarding the final photonic state in a multi-electron emission situation, in which more than one electron emits high-harmonic photons.

We have also studied the Wigner function as a possible physical observable in which absorption phenomena could be measured, but the main problem with respect to the obtained results lie on the fact that it is written in terms of its characteristic function, so if we apply the Fourier transform, four integrals have to be solved whose integration variables are t_1, t_2 and $z \equiv x_\varphi, x_{\bar{\varphi}}$. Hence, further work can be done in the search of other physical observables where quantum signatures could be imprinted, or also in other formulations of the Wigner function that can lead to simpler approaches.

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